Pattern Formation as a Signature of Quantum Degeneracy in a Cold Exciton System

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The development of a Turing instability to a spatially modulated state in a photoexcited electron-hole system is proposed as a novel signature of exciton Bose statistics. We show that such an instability, which is driven by kinetics of exciton formation, can result from stimulated processes that build up near quantum degeneracy. In the spatially uniform 2d electron-hole system, the instability leads to a triangular lattice pattern while, at an electron-hole interface, a periodic 1d pattern develops. We analyze the mechanism of wavelength selection, and show that the transition is abrupt (type I) for the uniform 2d system, and continuous (type II) for the electron-hole interface.

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Recently, striking spatial photoluminescence (PL) patterns [1, 2, 3, 4, 5], which span macroscopic scales in excess of $100\,\mu$m, have been observed in photoexcited AlGaAs/GaAs quantum well (QW) structures. In addition to concentric rings and 'bright spots', the electron-hole system exhibits an abrupt transition at ca. $2\,$K in which the outermost ring 'fragments' into regularly spaced beads of high PL intensity [1, 3]. While the gross features of PL have been explained within classical framework, attributing the internal rings to nonradiative exciton transport and cooling [1] and the outermost rings and 'bright spots' to macroscopic charge separation [3, 4, 5], the origin of the instability remains unidentified.

Spatially modulated exciton density is not to be expected in QW system designed so that excitons interact repulsively as electric dipoles [1] and thus do not form droplets [6]. The macroscopic character of ring fragments, of $10-30\,\mu$m in size and containing about 10^4 excitons each, and the abrupt temperature dependence, call for an explanation involving a symmetry-breaking instability of a homogeneous state to a patterned state. Such behavior is reminiscent of the instability predicted by Alan Turing [7] to occur in a reaction-diffusion system. The Turing instability is known to occur in certain chemical reactions [8, 9, 10], and is also believed to be relevant for pattern formation in biological systems [11].

In this work we propose a mechanism, based on the kinetics of exciton formation from optically excited electrons and holes, that can lead to instability in the exciton system. Interestingly, these kinetic effects become especially strong in the regime near *exciton quantum degeneracy*, due to stimulated enhancement of the electron-hole binding rate. The transition to a state with a spatially modulated exciton density reveals itself in the spatial PL pattern, and presents a directly observable signature of degeneracy.

Although, in itself, an observation of an instability does not constitute unambiguous evidence for degeneracy, it may complement other manifestations discussed in the literature, such as changes in exciton recombination [12] and scattering [13] rates, in the PL spectrum [14], absorption [15] and PL angular distribution [16]. While linking the observed instability with degeneracy is premature, our main aim here is to present the Turing instability from a broader viewpoint, as a novel and quite general effect of quantum kinetics that can help to identify the regime of Bose-Einstein condensation (BEC).

Here we consider a transport theory [3] formulated in terms of electron, hole, and exciton densities $n_{e,h,x}$ obeying a system of coupled nonlinear diffusion equations:

$$
\begin{aligned}
\text{(e)} \quad & \partial_t n_e = D_e \nabla^2 n_e - w n_e n_h + J_e \\
\text{(h)} \quad & \partial_t n_h = D_h \nabla^2 n_h - w n_e n_h + J_h \\
\text{(x)} \quad & \partial_t n_x = D_x \nabla^2 n_x + w n_e n_h - \gamma n_x.
\end{aligned}
\tag{1}
$$

The nonlinear couplings account for exciton formation from free electron and hole binding. Here, no attempt has been made to describe the detailed and complicated density and temperature dependence of the physical parameters entering the model, nor to account for nonequilibrium exciton energy distribution and cooling due to phonon emission [13, 17]. Instead, we adopt a more phenomenological approach and assume that, as a result of cooling, the system can be described by an effective temperature, which leaves the densities $n_{e,h,x}$ as the only important hydrodynamical variables. The sources J_e and J_h in Eq. (1) describe the carrier photo-production, as well as the leakage current in the QW structure.

In general, one can expect the electron-hole binding rate w and, to a lesser extent, the exciton recombination rate γ to depend sensitively on the local exciton density n_x. Of the several mechanisms that could lead to such a dependence at low temperatures close to exciton degeneracy, perhaps the most important in the present context involve stimulated electron-hole binding processes mediated by phonons. These processes enhance the binding rate by a factor $f = 1 + N_E^{eq}$, where N_E^{eq} denotes the occupation of exciton states. In thermal equilibrium, and at low temperatures, one can ignore the reverse processes

of exciton dissociation: The binding energy, carried away by phonons, is much larger than $k_B T$.

For a degenerate exciton gas with $N_{E=0} > 1$, the dominant process involves scattering into the ground state and the stimulated enhancement factor is expressed as

$$f = e^u, \quad u \equiv \frac{n_x}{n_0(T)}, \quad n_0(T) = \frac{2g m_x k_B T}{\pi \hbar^2}. \quad (2)$$

Here $m_x \simeq 0.21 m_0$ represents the exciton mass, and g denotes the degeneracy (in the indirect exciton system, the exchange interaction is extremely weak, and $g = 4$). Equivalently, when reparameterized through its dependence on temperature, $u \equiv T_0/T$ where $T_0 = \left(\pi \hbar^2/2g m_x k_B\right) n_x$ is the degeneracy temperature. At $T \sim T_0$ (equivalently $n_x \sim n_0$), there is a crossover from classical to quantum Bose-Einstein statistics, and the stimulated enhancement factor f increases sharply.

Qualitatively, the stimulated transition mechanism for hydrodynamic instability can be understood as follows: A local fluctuation in the exciton density leads to an increase in the stimulated electron-hole binding rate. The associated depletion of the local carrier concentration causes neighboring carriers to stream towards the point of fluctuation presenting a mechanism of positive feedback. The wavelength, determined by the most unstable harmonic of the density, characterizes the lengthscale of spatial modulation in the nonuniform state.

Before turning to the analysis of instability, it is useful to discuss intrinsic constraints on the dynamics (1) due to electric charge and particle number conservation. These are obtained by considering the linear combinations $(e) - (h)$, $(e) + (h) + 2(x)$ of the transport equations (1). In both cases, the nonlinear term drops out and one obtains linear equations

$$\hat{L}_e n_e - \hat{L}_h n_h = J_e - J_h \quad (3)$$

$$\hat{L}_e n_e + \hat{L}_h n_h + 2\hat{L}_x n_x + 2\gamma n_x = J_e + J_h \quad (4)$$

with $\hat{L}_{e(h,x)} = \partial_t - D_{e(h,x)} \nabla^2$. Note that, since the origin of the relations (3,4) is routed in conservation laws, they are robust and insensitive to the exact form of the electron-hole binding term.

Initially, let us consider a system in which the sources are constant $J_e(\mathbf{r}) = J_h(\mathbf{r}) \equiv J$, realized by a spatially extended photoexcitation. In this case, ignoring the dependence of the recombination rate γ on density, we have

$$\bar{n}_x = J/\gamma, \quad \bar{n}_{e,h} = (J/w(\bar{n}_x))^{1/2}. \quad (5)$$

The stability of the system can be assessed by linearizing Eqs. (1) about the uniform solution (5) with a harmonic modulation $\delta n_{e,h,x} \propto e^{\lambda t} e^{i\mathbf{k}\cdot\mathbf{r}}$. Using (3,4) and writing $\hat{L}_e \delta n_e = \hat{L}_h \delta n_h = -\hat{L}_x \delta n_x - \gamma \delta n_x$, one can express $\delta n_{h,x}$ in terms of δn_e and obtain

$$L_e(\lambda, \mathbf{k}) + \gamma \frac{\bar{n}_x}{\bar{n}_e} \left(1 + \frac{L_e(\lambda, \mathbf{k})}{L_h(\lambda, \mathbf{k})}\right) = \frac{\gamma u L_e(\lambda, \mathbf{k})}{L_x(\lambda, \mathbf{k}) + \gamma} \quad (6)$$

where $u = d\ln w/d\ln \bar{n}_x$ is evaluated at the steady state (5), and $L_{e(h,x)}(\lambda, \mathbf{k}) = \lambda + D_{e(h,x)} \mathbf{k}^2$. Solving Eq. (6), one obtains the growth rate dispersion $\lambda(\mathbf{k})$ from which one can infer from the stability criterion, $\text{Re}\,\lambda < 0$, that the system becomes unstable when

$$(\mathbf{k}\ell_x)^2 + r = u \frac{(\mathbf{k}\ell_x)^2}{1 + (\mathbf{k}\ell_x)^2}, \quad (7)$$

where $\ell_x = \sqrt{D_x/\gamma}$ denotes the exciton diffusion length and $r = D_x(D_e^{-1} + D_h^{-1})\bar{n}_x/\bar{n}_e$. Eq. (7) has solutions if

$$u \equiv \frac{d\ln w}{d\ln \bar{n}_x} \geq u_c = \left(1 + r^{1/2}\right)^2 \quad (8)$$

as illustrated in Fig. 1. At $u = u_c$, we obtain the most unstable wavenumber $k_* = r^{1/4}/\ell_x$ selected by competition of the stimulated binding and diffusion processes.

The binding rate $w(n_x)$ builds up near degeneracy due to the growth of stimulated processes, leading to instability at temperatures approaching T_{BEC}. (For a weakly interacting Bose gas the transport coefficients, and thus the constant r, are practically insensitive to the degree of exciton degeneracy at $T > T_{\text{BEC}}$.)
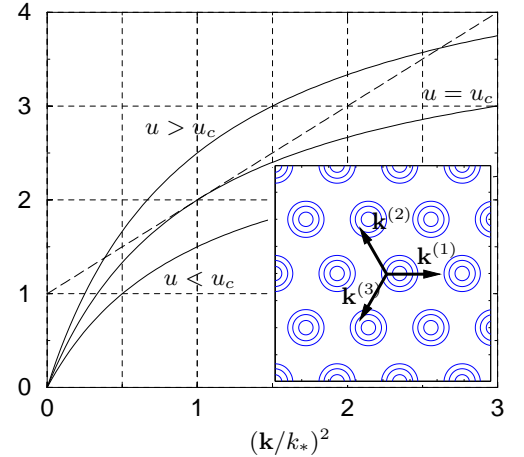

FIG. 1: Graphical solution of Eq. (7) that selects the most unstable wavelength. *Inset:* The 3-fold symmetric star of wavevectors describing the modulation near the instability threshold and the corresponding triangular pattern of exciton density variation.

To what extent are these results insensitive to the origin of the nonlinearity in the binding rate? If enhanced by intraband Auger processes, which transfer the binding energy released in exciton formation to other excitons, one expects the binding rate w to scale linearly with local exciton density, viz. $w(n_x) = w_0(1 + n_x/\tilde{n}_0)$, where $\tilde{n}_0$ denotes some constant involving a ratio of the two-body and three-body cross-section of the electron and hole in the presence of excitons. Crucially, in this case, the left hand side of Eq. (8) is bounded by unity, while the right hand side is in excess. Therefore, at least over the parameter range considered here, one can infer that a simple

linear scaling of the binding rate with density does not lead to instability. Indeed, the instability may be used to discriminate against certain mechanisms in the kinetics of exciton formation.

Turning to the discussion of the spatial pattern resulting from the instability, we note that the wavevector selection determines its modulus, but not direction. At threshold $u = u_c$ all modes with $|\mathbf{k}| = k_*$ become unstable simultaneously. The resulting 2d density distribution can be found by considering the effect of mixing different harmonics due to higher order terms in (1) expanded in $\delta n_{e,x}$ about the uniform state. Since these equations contain quadratic terms, the favored combination of harmonics is a 3-fold symmetric star $\mathbf{k}^{(j)} = k_*(\cos(\frac{2\pi}{3}j+\theta), \sin(\frac{2\pi}{3}j+\theta))$, $j = 1, 2, 3$, with the parameter θ describing the degeneracy with respect to 2d rotations. This leads to a density distribution $\delta n \propto \sum_j e^{\pm \mathbf{k}_j \cdot \mathbf{r}}$ with maxima arranged in a triangular lattice.

On symmetry grounds, since the triangular lattice pattern is stabilized by quadratic terms, the mean field analysis predicts that the transition to the modulated state in this case is abrupt, of a type I kind. Indeed, the triangular lattice geometry arises in various 2d pattern selection problems, from Bénard convection cells [18] to the mixed state of type II superconductors [19].

To explore the application of these ideas to the 1d modulation seen in exciton rings [1, 3], one must first determine the profile of the uniform distribution. The rings represent an interface between regions populated by electrons and holes at which they bind to form excitons. The steady state is maintained by a constant flux of carriers arriving at the interface. The parameter regime which is both relevant and simple to analyze is that of long exciton lifetime γ^{-1} where the diffusion length ℓ_x exceeds the range of the electron and hole profile overlap. In this case, approximating the source of excitons by a straight line $c\delta(x)$, where c is the total carrier flux and x is the coordinate normal to the interface, the exciton density profile is given by $(c\ell_x/2D_x)e^{-|x|/\ell_x}$. Accordingly, one can seek the electron and hole profile treating $w(n_x)$ as constant and restoring its dependence on n_x later when turning to the instability. The profiles can be inferred from two coupled nonlinear diffusion equations

$$D_{e(h)}\partial_x^2 n_{e(h)} = w n_e n_h, \qquad (9)$$

with the boundary condition: $D_e \partial_x n_e^e|_{\pm\infty} = \pm c\,\theta(\pm x)$. From Eq. (3) one obtains $D_e n_e - D_h n_h = cx$ which allows the elimination of n_h. Applying the rescaling $n_{e(h)} = c\ell\, g_{e(h)}/D_{e(h)}$, where $\ell = (D_e D_h/wc)^{1/3}$, one obtains

$$\partial_{\tilde{x}}^2\, g_e = g_e(g_e - \tilde{x}), \quad \tilde{x} \equiv x/\ell. \qquad (10)$$

From the rescaling one can infer that the electron and hole profiles overlap in a range of width $\ell \sim c^{-1/3}$ while

$$g_e(|x| \gg \ell) = \tilde{x}\,\theta(\tilde{x}) + \mathcal{O}\left(|\tilde{x}|^{-1/4}e^{-2|\tilde{x}|^{3/2}/3}\right).$$
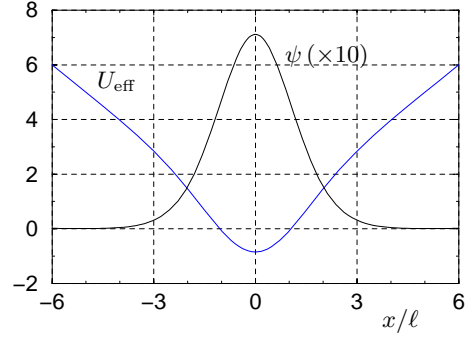

FIG. 2: Numerical solution of the (normalized) bound state wavefunction $\psi(x)$ representing fluctuation in the electron density, together with the effective potential $U_{\text{eff}} = \bar{g}_e + \bar{g}_h - a_0\,\bar{g}_e\bar{g}_h$ of the Schrödinger-like equation (11).

Although Eqs. (9) are nonlinear, their diffusive character does not straightforwardly admit a spatial instability: A fluctuation in the position of the interface initiates an increased electron-hole flux which, in time, restores the uniform distribution. However, if one restores the dependence of the binding rate on exciton density, the same mechanism of positive feedback which characterized the instability in the uniform system becomes active. To explore the instability, one may again expand linearly in fluctuations around the spatially uniform solution, $g_e(x, y) = \bar{g}_e(x) + \delta g_e(x)\, e^{iky}$ (similarly g_h and g_x), where y is the coordinate along the interface and $\bar{g}_e(x)$ denotes the uniform profile obtained from Eq. (10). With $\ell_x \gg \ell$, the exciton density remains roughly uniform over the electron-hole interface. Denoting this value by $\bar{n}_x(0)$, in the vicinity of the interface, one may again develop the linear expansion $w[n_x] \simeq w[\bar{n}_x(0)]\,(1 + u\delta n_x/\bar{n}_x(0))$ where, as before, $u = d\ln w/d\ln n_x$. Noting that Eq. (3) enforces the steady state condition $\delta g_e = \delta g_h$, a linearisation of Eqs.(1) obtains the Schrödinger-like equation

$$\left[-\partial_{\tilde{x}}^2 + (\ell k)^2 + \bar{g}_e + \bar{g}_h\right]\delta g_e + \frac{u}{\bar{g}_x(0)}\bar{g}_e\bar{g}_h\,\delta g_x = 0, \quad (11)$$

together with the condition on the Fourier components,

$$\delta g_x(q) = -\delta g_e(q)\left(1 - \frac{1}{Q^2 + (q\ell_x)^2}\right), \qquad (12)$$

with $Q^2 \equiv (k\ell_x)^2 + 1$. Now, since the product $\bar{g}_e\bar{g}_h$ is strongly peaked around the interface, the typical contribution from the last term in (11) arises from Fourier elements $q\ell \sim 1$. Then, with $\ell_x \gg \ell$, the second contribution to $\delta g_x(q)$ can be treated as a small perturbation on the first and, to leading order, neglected, i.e. $\delta g_x \simeq -\delta g_e$. In this approximation, the most unstable mode occurs at $k = 0$. Qualitatively, an increase in u will trigger an instability of the $k = 0$ mode at a critical value u_c when the linear equation first admits a non-zero solution for δg_e. At the critical point, the corresponding fluctuation in the electron density then acquires the profile of the (normalized) zero energy eigenstate $\psi(x)$. Numerically

one finds that the critical point for the instability occurs when $u_c/\bar{g}_x(0) = (2\ell/\ell_x)u_c \equiv a_0 \simeq 6.516$, while the corresponding solution $\psi(x)$ is shown in Fig. 2.

While the approximation above identifies an instability, a perturbative analysis of the k-dependent corrections implied by (12) reveals that the most unstable mode is spatially modulated. To the leading order of perturbation theory, an estimate of the shift of u_c obtains

$$\frac{\delta u_c}{u_c} = \frac{\ell}{a_0 a_1}\left((k\ell)^2 + \frac{a_0 a_2(Q)}{\ell_x^2 Q}\right)$$

where $a_1 = \int_{-\infty}^{\infty} dx\, \bar{g}_e(x)\bar{g}_h(x)\psi^2(x) \simeq 0.254\ell$, and

$$a_2(Q) = \frac{1}{2}\int_{-\infty}^{\infty} dxdx'\, \bar{g}_e(x)\bar{g}_h(x)\psi(x)e^{-Q|x-x'|/\ell_x}\psi(x').$$

With $\ell_x \gg \ell$, it will follow that $Q\ell \ll \ell_x$, and the latter takes the constant value $a_2 \simeq 0.461\ell^2$ independent of k. Finally, minimizing δu_c with respect to k, one finds that the instability occurs with a wavevector

$$k_c\ell_x \simeq \left(a_0 a_2 \ell_x/2\ell^3\right)^{1/3} \qquad (13)$$

implying a shift of u_c by $\delta u_c/u_c \sim (\ell/\ell_x)^{4/3}$. As a result, one can infer that the spatial modulation wavelength $\lambda_c \sim \ell^{1/3}\ell_x^{2/3}$ is typically larger than the electron-hole overlap ℓ, but smaller than ℓ_x. Finally, an expansion of the nonlinear equations to higher order in fluctuations shows that, below the transition (i.e. for $u > u_c$), the amplitude of the Fourier harmonic k_c grows as $(u - u_c)^{1/2}$.
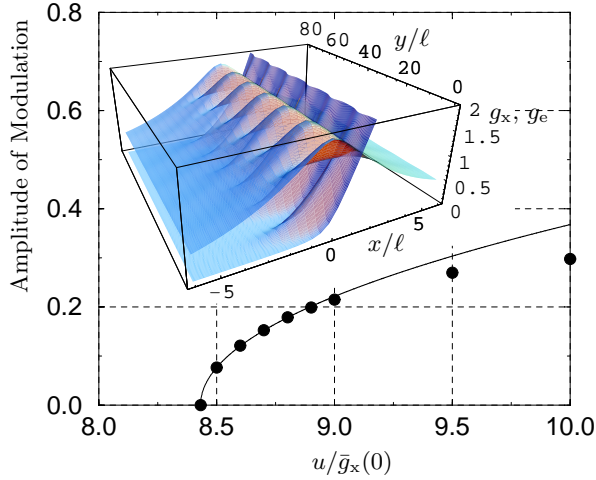

FIG. 3: Amplitude of the spatial modulation of the exciton density g_x along the electron-hole interface $x = 0$ as a function of the control parameter $u/\bar{g}_x(0)$ as determined from the numerical solution of the nonlinear transport equation (14). A fit of the data to a square root dependence $\sim (u - u_c)^{1/2}$ is shown. Inset: Dimensionless electron g_e and exciton g_x densities for $u/\bar{g}_x(0) = 9.5$ and $\ell_x/\ell = 4$. Here periodic boundary conditions are imposed along the interface.

Once the instability is strongly developed (or when $\ell_x \lesssim \ell$) the linear stability analysis above becomes unreliable. Here one must turn to numerics. Having in mind

the mechanism of stimulated scattering, Fig. 3 shows the results of a numerical analysis of the dimensionless nonlinear steady-state equation

$$\nabla_{\tilde{x}}^2\, g_e = \exp\left(\frac{u}{\bar{g}_x(0)}\delta g_x\right)g_e(g_e - \tilde{x}), \qquad (14)$$

where, using Eq. (12), $\delta g_x \equiv g_x - \bar{g}_x$ depends non-locally on $\delta g_e \equiv g_e - \bar{g}_e$ through the linear relation

$$\delta g_x(\mathbf{x}) = -\delta g_e(\mathbf{x}) + \int \frac{d^2\mathbf{x}'}{2\pi\ell_x^2}K_0\left(\frac{|\mathbf{x}-\mathbf{x}'|}{\ell_x}\right)\delta g_e(\mathbf{x}'),$$

and K_0 denotes the modified Bessel function. Although (as in the experimental ring geometry) the modulation is constrained by the periodic boundary conditions imposed along the direction parallel to the interface, the critical wavenumber k_c lies close to that predicted by Eq. (13). Similarly, the constraint leads to a value of $u_c/\bar{g}_x(0)$ a little in excess of that predicted by the linear stability analysis. Finally, the amplitude of the instability confirms the square root dependence on $(u - u_c)$ predicted by perturbation theory.

In summary, we have shown that the realization of quantum degeneracy in a cold electron/hole-exciton system is signalled by the development of a spatial density modulation. Although our discussion is motivated by the photoexcited CQW system in which electrons and holes are spatially separated, the mechanism is quite generic applying also to geometries where the electron and hole sources $J_{e,h}$ have a spatially independent profile. By contrast, the instability mechanism appears to depend sensitively on there being a *strongly* nonlinear dependence of the electron-hole binding rate on the exciton density pointing to the importance of stimulated scattering.

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